Hidden long range order in Heisenberg Kagomé antiferromagnets

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We give a physical picture of the low-energy sector of the spin 1/2 Heisenberg Kagomé antiferromagnet (KAF). It is shown that Kagomé lattice can be presented as a set of stars which are arranged in a triangular lattice and contain 12 spins. Each of these stars has two degenerate singlet ground states which can be considered in terms of pseudospin. As a result of interaction between stars we get Hamiltonian of the Ising ferromagnet in magnetic field. So in contrast to the common view there is a long range order in KAF consisting of definite singlet states of the stars.

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In spite of numerous theoretical and experimental studies in the last decade, some magnetic properties of Kagomé antiferromagnets (KAFs) remain open problems. Experiments revealed unusual low-temperature behavior of the specific heat and magnetic susceptibility in Kagomé -like compounds. For example specific heat C measurements in $SrCr_{9p}Ga_{12-9p}O_{19}$ ($S = 3/2$ Kagomé material) have shown that there is a peak at $T \approx 5$ K, $C \propto T^2$ at $T \lesssim 5$ K and it appears to be practically independent of magnetic field up to 12 T [1].

Some of the experimental findings are in agreement with the results of numerical finite cluster investigations [2, 3, 4, 5]. They reveal a gap separating the ground state from the upper triplet levels and a band of nonmagnetic singlet excitations with a very small or zero gap inside the triplet gap. The number of states in the band increases with the number of sites N as α^N. For samples with up to 36 sites $\alpha = 1.15$ and 1.18 for the even and odd N, respectively [3]. This wealth of low-lying singlet excitations explains the peak of specific heat at low temperature and its independence of the magnetic field.

However the origin of this band as well as the nature of the ground state were unclear until now. Previous exact diagonalization studies [4, 6] reveal exponential decay of the spin-spin and dimer-dimer correlation functions. So the point of view that KAF is a spin liquid is widely accepted now [3, 4, 5, 6, 7, 8, 9, 10]. It seems the best candidate for description of KAF low-energy properties is a quantum dimer model [5, 7, 11]. It should be mentioned a certain recent success in this field. In the paper [12] a spin 1/2 Kagomé lattice is considered as a set of interactive triangles with a spin in each apex. It was suggested there to work in the subspace where the total spin of each triangle is 1/2 (short range RVB states (SRRVB)) investigating low-lying excitations. It was shown that low-energy spectrum obtained with SRRVB on the samples with up to 36 cites and the number of singlet excitations in the band coincide with the results of exact diagonalization. Meanwhile a further development of this approach is required to get a full physical description of KAF.

Another types of frustrated magnets which possess a similar behavior as KAF and have many singlet states inside the triplet gap are pyrochlore [13] and CaV_4O_9 [14]. Recently it was suggested a model of frustrated antiferromagnet which low-energy properties can be generic for these compounds as well as for KAF [15]. Weakly interactive plaquettes in the square lattice were considered there. Each plaquette has two almost degenerate singlet ground states, so a band of singlet excitations arises if the inter-plaquette interaction is taken into account. It is shown that there is a quantum phase transition in the model at a critical value of frustration separating a disorder plaquette phase and a columnar dimer one. In the proximity of this transition the specific heat has a low-temperature peak below which it possesses a power low temperature dependence.

In this paper we show that such a behavior is do relevant for spin 1/2 KAF. It is proposed to consider a Kagomé lattice as a set of stars with 12 spins arranged in a triangular lattice (see Fig. 1). A star has two degenerate singlet ground states. Interaction between stars leads to the band of low-lying excitations which number increases as $2^{N/12} \approx 1.06^N$. It is demonstrated that this interaction can be considered as a perturbation in the low-energy sector. As a result we get a model of the Ising ferromagnet in effective magnetic field where these degenerate states are described in terms of two projections of pseudospins 1/2. So it is shown that in contrast to the common view there is a hidden long range order in KAF which consists of definite singlet states of the stars. This picture should be relevant also for KAFs with larger values of spin.

We start with the Hamiltonian of the spin 1/2 Kagomé Heisenberg antiferromagnet:

$$\mathcal{H}_0 = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{(i,j)} \mathbf{S}_i \mathbf{S}_j, \tag{1}$$

where $\langle i,j \rangle$ and (i,j) denote nearest and next-nearest neighbors on the Kagomé lattice shown in Fig. 1, respectively. The case of $|J_2| \ll J_1$ is considered in this paper. We discuss a possibility of both signs of next-nearest-neighbor interaction — ferromagnet and antifer-

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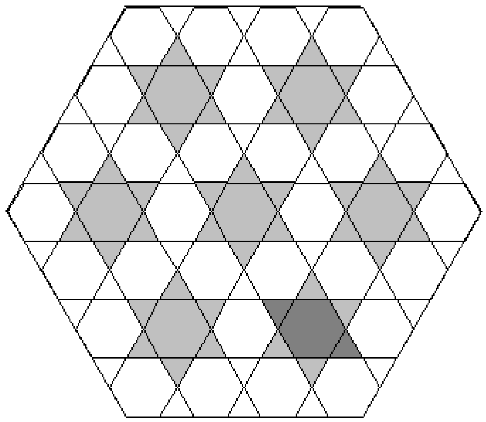

FIG. 1: Kagomé lattice (KL). There is a spin in each lattice site. KL can be considered as a set of stars arranged in a triangular lattice. Each star contains 12 spins. An unit cell is also presented (dark region). There are four unit cells per a star.

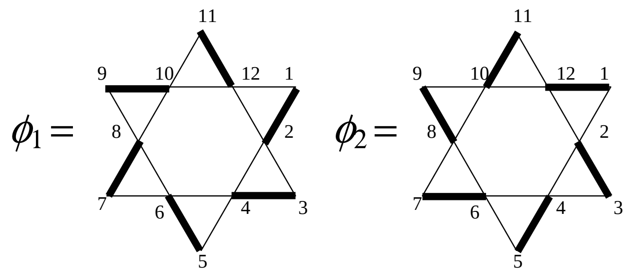

FIG. 2: Schematic representation of a star's two singlet ground states wave functions ϕ_1 and ϕ_2. A bold line denotes the singlet state of two neighboring spins, i.e. $(|\uparrow\rangle_i|\downarrow\rangle_j - |\downarrow\rangle_i|\uparrow\rangle_j)/\sqrt{2}$.

romagnet one. As is shown below, in spite of the smallness the second term in Eq. (1) can be of importance for the low-energy properties.

Kagomé lattice can be presented as a set of stars arranged in a triangular lattice (see Fig. 1). To begin with we neglect interaction between stars and put $J_2 = 0$ in Eq. (1). A star is a system of 12 spins. Let us consider its properties in detail. The symmetry group contains 6 rotations and reflections with respect to 6 lines passing through the center. There are two degenerate singlet ground states ϕ_1 and ϕ_2 which differ each other by symmetry. Their wave functions are shown schematically in Fig. 2 where a bold line represents the singlet state of the corresponding two spins, i.e. $(|\uparrow\rangle_i|\downarrow\rangle_j - |\downarrow\rangle_i|\uparrow\rangle_j)/\sqrt{2}$. Evidently ϕ_1 and ϕ_2 are invariant with respect to rotations of the star and they transform to each other under reflections. They contain six singlets each having the energy $-3/4J_1$. One can show that interaction between singlets makes no contribution to the energy of the ground states which is consequently equal to $-4.5J_1$. We have obtained numerically that there is a gap of the value approximately $0.26J_1$ which separates the ground states and the lower triplet levels in the star.

It should be pointed out that ϕ_1 and ϕ_2 are not orthogonal: the scalar product of these two functions is $(\phi_1\phi_2) = 1/32$. So in the following we will use two orthonormalized combinations:

$$\Psi_1 = \frac{1}{\sqrt{2+1/16}}(\phi_1 + \phi_2), \tag{2}$$

$$\Psi_2 = \frac{1}{\sqrt{2-1/16}}(\phi_1 - \phi_2). \tag{3}$$

Because ϕ_1 and ϕ_2 are invariant under rotations and transform to each other with reflections, Ψ_1 is invariant under all the symmetry group transformations and Ψ_2 is invariant under rotations and it changes the sign with reflections.

We turn now to consideration of the interaction between two nearest stars still neglecting the second term in Eq. (1). Initially there are four degenerate ground states with energy $E_0 = -9J_1$ and wave functions $\{\Psi^{(1)}_{n_1}\Psi^{(2)}_{n_2}\}$ ($n_i = 1, 2$), where upper index labels the stars. As it is seen from Fig. 3, the interaction energy has the form:

$$V = J_1(\mathbf{S}^{(1)}_1\mathbf{S}^{(2)}_1 + \mathbf{S}^{(1)}_3\mathbf{S}^{(2)}_3). \tag{4}$$

Let us consider V as a perturbation. According to the standard theory [16] one have to solve a secular equation to find the first correction to the energy. In our case there are four equations and the corresponding matrix elements are given by

$$H_{n_1n_2;k_1k_2} = V_{n_1n_2;k_1k_2} + \sum_{m_1,m_2} \frac{V_{n_1n_2;m_1m_2}V_{m_1m_2;k_1k_2}}{E_0 - E_{m_1} - E_{m_2}}, \tag{5}$$

where $V_{n_1n_2;k_1k_2} = \langle\Psi^{(1)}_{n_1}\Psi^{(2)}_{n_2} \mid V \mid \Psi^{(1)}_{k_1}\Psi^{(2)}_{k_2}\rangle$, $n_i, k_i = 1, 2$ and indexes m_1 and m_2 denote excited levels of the first and the second star, respectively. Obviously the first term in Eq. (5) is zero and the second one can be represented as follows:

$$H_{n_1n_2;k_1k_2} = -i\int_0^\infty dt e^{-\delta t + iE_0 t} \langle\Psi^{(1)}_{n_1}\Psi^{(2)}_{n_2} \mid V e^{-it(H_{01}+H_{02})} V \mid \Psi^{(1)}_{k_1}\Psi^{(2)}_{k_2}\rangle, \tag{6}$$

where H_{0i} are Hamiltonians of the corresponding stars. Using the symmetry of the functions ϕ_1, ϕ_2, Ψ_1 and Ψ_2 discussed above one can show that only diagonal elements (i.e. $n_1 = k_1$, $n_2 = k_2$) in Eq. (6) are nonzero. We have calculated them with a very high precision by expansion of the operator exponent up to the power 130. The results can be represented in the following form:

$$H_{11;11} = -a_1 + a_2 - a_3, \tag{7a}$$

$$H_{12;12} = -a_1 + a_3, \tag{7b}$$

$$H_{21;21} = -a_1 + a_3, \tag{7c}$$

$$H_{22;22} = -a_1 - a_2 - a_3, \tag{7d}$$

where $a_1 = 0.256J_1$, $a_2 = 0.015J_1$ and $a_3 = 0.0027J_1$. So the interaction shifts all the levels on the value $-a_1$ and lifts their degeneracy. Constants a_2 and a_3 in Eqs. (7) determine the levels splitting. All corrections are small enough and one can consider interaction Eq. (4) between stars as a perturbation at low energies. We restrict ourself with this precision here and don't consider triplet states.

So KAF appears to be a set of two-levels interacting systems and one can naturally represent the low-energy singlet sector of Hilbert space in terms of pseudospins: $|\uparrow\rangle = \Psi_2$ and $|\downarrow\rangle = \Psi_1$. It is seen from Eqs. (7) that in these terms the interaction between stars is described by the Hamiltonian of Ising ferromagnet in the external magnetic field:

$$\mathcal{H} = -\mathcal{J} \sum_{\langle i,j \rangle} s_i^z s_j^z - h \sum_i s_i^z, \tag{8}$$

where $\langle i,j \rangle$ labes now nearest-neighbor pseudospins, arranged in a triangular lattice formed by the stars, $\mathcal{J} = 4a_3 = 0.011J_1$ and $h = 6a_2 = 0.092J_1$. We also omit a constant in Eq. (8) which is equal to $-0.439J_1N$. It should be stressed that within our precision Hamiltonian Eq. (8) is an exact mapping of the original Heisenberg model in the low-energy sector (excitation energy $\omega \sim \mathcal{J}$). So one can see from Eq. (8) that the ground state of KAF is that with all the stars in Ψ_2 states.

In fact we show existence of a long range order in KAF generated by singlets. This hidden order settles on the triangular star lattice and can be checked by inelastic neutron scattering: corresponding intensity for the singlet-triplet transitions should have periodicity in the reciprocal space corresponding to the original star lattice. This picture is similar to observed one in the case of the dimerised spin-Pairls compound $CuGeO_3$ [17]. Because low-energy physics in KAF is determined by singlets, our consideration should be relevant qualitatively also for KAFs with the larger values of spin.

We proceed with the discussion of the number of low-energy states in KAF. As each star has two singlet ground states and contains 12 spins the number of singlet excitations in the band is given by $2^{N/12} \approx 1.06^N$. Unfortunately there is no point to compare this result with that of the previous numerical work [3] discussed above because a very small samples ($N \leq 36$) were considered there.

Let us take into account the next-nearest-neighbor interactions. As is seen from Fig. 3 they can be divided on three parts: $\tilde{V}_1$, $\tilde{V}_2^{(1)}$ and $\tilde{V}_2^{(2)}$, where $\tilde{V}_1 = J_2(\mathbf{S}_1^{(1)}\mathbf{S}_2^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_3^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_2^{(2)})$ contributes to the inter-stars interaction and $\tilde{V}_2^{(1)}$ and $\tilde{V}_2^{(2)}$ contain 12 intrinsic next-nearest-neighbor interactions of the first and the second star, respectively. Considering now perturbation theory according to a sum of these three operators and that given by Eq. (4) we find that in addition to corrections presented in Eqs. (7) there are new ones proportional to J_2 from the first and the second terms in

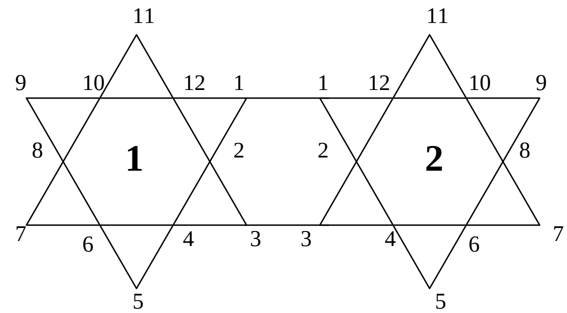

FIG. 3: There are the following interactions between each two stars: $V = J_1(\mathbf{S}_1^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_3^{(2)})$ and $\tilde{V}_1 = J_2(\mathbf{S}_1^{(1)}\mathbf{S}_2^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_3^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_2^{(2)})$, where upper indexes label the stars.

Eq. (5) given by $\tilde{V}_2^{(1)}$, $\tilde{V}_2^{(2)}$ and $\tilde{V}_1$, respectively. Using symmetry of functions ϕ_1 and ϕ_2 it can be shown that secular matrix Eq. (6) remains diagonal in this case. As a result calculations give for the values of "exchange" and "magnetic field" in the effective Hamiltonian Eq. (8):

$$\mathcal{J} = 0.011J_1 - 0.005J_2, \tag{9}$$
$$h = 0.092J_1 + 3.635J_2. \tag{10}$$

One can see from Eqs. (9) and (10) that the next-nearest interactions give a correction of the order of $|J_2|/J_1 \ll 1$ to the value $\mathcal{J}$ and their contribution to the "magnetic field" is considerable if $|J_2| \gtrsim 0.01J_1$. If $J_2 < 0$ (ferromagnet interaction) they could even change the sign of h. In the case of $h > 0$ the ground state of the Kagomé lattice is that with all stars in Ψ_2 states and if $h < 0$ all stars are in Ψ_1 states.

We could expect a logarithmic singularity of the specific heat C in the point $h = 0$ at the critical temperature T_c which is of the order of $\mathcal{J}$ and there should be a peak at $T \sim \mathcal{J}$ if $h \neq 0$. Specific heat decreases at $T \to 0$ as $e^{-(3\mathcal{J}+|h|)/T}$. So we don't get low-temperature behavior $C \propto T^2$ obtained in experiments [1]. It should be noted that such a law should exist if the energy of low-lying excitations $\epsilon_{\mathbf{q}}$ with wave vector $\mathbf{q}$ at $q \ll 1$ has the form $\epsilon_{\mathbf{q}} = cq^2 + \Delta$. Within the first order of perturbation theory considered in this paper interaction between stars is described by the Hamiltonian Eq. (8) of Ising ferromagnet in magnetic field which doesn't imply such a behavior of the low-energy spectrum. But the further orders could give some kind of anisotropy in Eq. (8) which leads to the necessary picture. This point will be considered in detail elsewhere.

It is appropriate to mention here a recent experiment on $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ [18] which is the only candidate for spin-1/2 Kagomé material by now. In spite of strong exchange $J_1 \sim 100$ K in this compound, there is no regime obtained for KAF with the larger values of spins has been achieved up to the temperature 1.8 K. In this respect we point here on a small scale of the dynamics in spin-1/2 KAF. According to our results the representa-

tive temperature is of the order of $0.01J_1$, so the region $T \lesssim 1$ K should be attained for this material.

In conclusion, we present a new insight of low-energy physics of spin 1/2 Kagomé antiferromagnet (KAF). The lattice can be presented as a set of stars which are arranged in a triangular lattice and contain 12 spins (see Fig. 1). Each star has two degenerate singlet ground states with different symmetry. It is shown that interaction between the stars leads to the band of singlet excitations and can be considered as a perturbation in the low-energy sector. We demonstrate the existence of a long range order in KAF on the triangular star lattice which is generated by singlets and can be detected in particular in experiments on inelastic neutron scattering. This physical picture should be relevant also for KAFs with larger spin values.

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